

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
22 February 2001 (22.02.2001)

PCT

(10) International Publication Number  
**WO 01/12148 A1**

(51) International Patent Classification<sup>7</sup>: **A61K 7/48**

(21) International Application Number: **PCT/EP00/06796**

(22) International Filing Date: **14 July 2000 (14.07.2000)**

(25) Filing Language: **English**

(26) Publication Language: **English**

(30) Priority Data:  
**60/148,774** **13 August 1999 (13.08.1999)** **US**

(71) Applicant (for AE, AG, AU, BB, BZ, CA, CY, GB, GD, GH, GM, IE, IL, KE, LC, LK, LS, MN, MW, NZ, SD, SG, SL, SZ, TT, TZ, UG, ZA, ZW only): **UNILEVER PLC** [GB/GB];  
Unilever House, Blackfriars, London EC4P 4BQ (GB).

(71) Applicant (for all designated States except AE, AG, AU, BB, BZ, CA, CY, GB, GD, GH, GM, IE, IL, IN, KE, LC, LK, LS, MN, MW, NZ, SD, SG, SL, SZ, TT, TZ, UG, ZA, ZW): **UNILEVER NV** [NL/NL]; Weena 455, NL-3013 AL Rotterdam (NL).

(71) Applicant (for IN only): **HINDUSTAN LEVER LIMITED** [IN/IN]; Hindustan Lever House, 165/166 Backbay Reclamation, Maharashtra, 400 020 Mumbai (IN).

(72) Inventors: **SLAVTCHEFF, Craig, Stephen**; Unilever Home & Personal Care USA, 40 Merritt Boulevard, Trumbull, CT 06611 (US). **ZNAIDEN, Alexander, Paul**; Nippon Lever B.V., Utsunomiya Factory/Innovation Centre, 38 Haga-dai, Haga-cho, Haga-gun, Tochigi 321-33

(JP). **INDURSKY, Michael**; Unilever Home & Personal Care USA, 33 Benedict Place, Greenwich, CT 06830 (US). **CROTTY, Brian, Andrew**; Unilever Home & Personal Care USA, 40 Merritt Boulevard, Trumbull, CT 06611 (US).

(74) Agents: **ROTS, Maria, Johanna, Francisca et al.**; Unilever Plc. Patent Department, Colworth House, Sharnbrook, Bedford, Bedfordshire MK44 1LQ (GB).

(81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW.

(84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

**Published:**

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: **COSMETIC STRIPS WITH LIQUID CRYSTAL TEMPERATURE DEPENDENT COLOR CHANGE**

(57) Abstract: An adhesive cosmetic strip is provided which includes a flexible water-insoluble substrate, an adhesive composition deposited onto the substrate, a liquid crystal thermochromic substance, and an agent interactive with water to ensure a temperature rise of at least 2°C in the strip. The thermochromic substance may be impregnated into the substrate or dispersed within the adhesive composition. When applied to the skin, the strip is warmed by a reaction between water and an exothermic or endothermic agent held within the strip. Moisture within the skin or externally applied water penetrates the strip and will react with the agent to cause an increase in temperature. Subsequently this increase in temperature will induce a color change in the thermochromic substance.

- 1 -

COSMETIC STRIPS WITH LIQUID CRYSTAL  
TEMPERATURE DEPENDENT COLOR CHANGE

BACKGROUND OF THE INVENTION

5

Field of the Invention

The invention concerns cosmetic dermal strips or patches which provide consumers with a color change indicator as a sensorial signal.

10

The Related Art

Cosmetics are often provided with consumer perceivable sensorial signals. Most common of these signals are fragrances. Pleasing odor is often the single most important attribute inducing re-purchase by a consumer. Other sensorial attributes are also significant in cosmetic chemistry. Skinfeel of a product is highly important. Creams, lotions, gels and pastes often are judged for their efficacy by the tactility of their feel. Silky, non-residue leaving cosmetics are much preferred over tacky ones, and the consumer may relate those aesthetics to actual pharmacological performance.

25

Sometimes the sensorial attribute is that of temperature. Coolness is imparted to toothpastes and aftershave lotions through the presence of camphor, menthol or menthol derivatives such as menthol lactate. Some formulations signal efficacy through a temperature increase. Inclusion

30

- 2 -

of capsaicin, an alkaloid extracted from capsicum, gives a brief temperature rise sensation to the human neural system.

Dermal patches or strips have recently become popular as delivery vehicle systems for cosmetic compositions. For instance, WO 98/42303 (Crotty et al.) describes a dry-to-the-touch keratotic plug remover strip. Upon wetting, the strip turns tacky and mobile. This product is placed on the bridge of the nose or other areas of the face requiring keratotic plug removal. Within a short time period, water evaporates from the wetted adhesive forming a dry film. The consumer must then peel the film from the face along with unwanted plugs bonded thereto. Amounts of water applied by the consumer may vary. Drying times are therefore also variable. A sensorial signal would be helpful for the consumer to know when to begin the peel removal.

Exothermic and endothermic reactions are other sources of temperature signaling. U.S. Patent 5,861,440 (Gohla et al.) reports use of sugars, especially xylitol, for inducing a cooling sensation when contacted with water. Generation of exothermic heat is reported in U.S. Patent 4,379,143 (Sherry et al.), U.S. Patent 4,626,550 (Hertzenberg), U.S. Patent 4,362,715 (Strianse et al.) and U.S. Patent 3,250,680 (Menkart et al.). Each of these patents employ an aluminosilicate interacting with water to release momentary heat.

Other types of sensorial signals have been sought for incorporation into cosmetics. The signals should either

- 3 -

provide an independently new effect or complement those which have traditionally been employed.

Accordingly, it is an object of the present invention to  
5 provide cosmetic products, especially dermal strips or patches with a new sensorial signal.

Still another object of the present invention is to provide cosmetic products, especially dermal strips or patches with  
10 a sensorial signal which may augment other sensory or emotive aesthetics of such products.

Yet another object of the present invention is to provide cosmetic products, especially dermal strips or patches which  
15 include a timing mechanism for application, rub-in or removal of the product from a consumer's skin.

These and other objects of the present invention will become more readily apparent from consideration of the following  
20 summary and detailed discussion.

#### **SUMMARY OF THE INVENTION**

An adhesive cosmetic strip is provided for placement on skin  
25 which includes:

- (i) a flexible water-insoluble substrate;
- (ii) an adhesive composition deposited onto the substrate;
- (iii) an agent interactive with water present in a  
30 sufficient quantity to induce a change of temperature in the strip of at least 2°C; and

- 4 -

(iv) a liquid crystal thermochromic substance incorporated into at least one of the substrate or adhesive composition, the thermochromic substance changing color in response to the change of temperature.

#### DETAILED DESCRIPTION OF THE INVENTION

Now a visual sensorial signal has been found for use with dermal strips or patches. The invention is based upon the use of liquid crystal thermochromic substances whose color changes in response to a change in temperature. The substances may either be incorporated into the flexible substrate structurally supporting the strip or patch or may be formulated with the adhesive.

Liquid crystal thermochromic substances ordinarily are colorless (essentially transparent) except when in a specific temperature range, at which time they become visible. Often mixtures of these substances are employed to change different colors responsive to changes in temperature. They may be cholesteric or chiral nematic liquid crystals. Commercially the substances are available from Hallcrest of Glenview, Illinois. Hallcrest has developed a silk-screenable liquid crystal material which is called a microencapsulated chiral nematic with vivid colors. Since the liquid crystal scatters light, it thus requires a generally dark background to be most visible. It changes through its liquid crystal visible color spectrum of clear to red to green to blue to purple to clear (as heated) or the reverse color sequence (when cooled). Temperature below

- 5 -

its design temperature range, the liquid crystal substance is essentially transparent. Similarly, the temperature above its design temperature range, the liquid crystal material is also essentially transparent. Therefore, the liquid crystal material is only visible when at a temperature within the desired temperature range. Typical liquid crystals are cholesteryl ester carbonates or chiral nematic (non-sterol) aryl compounds. Particularly useful are (2-methylbutyl) phenol 4-alkyl(oxy)benzoates. Hallcrest provides microencapsulated thermochromic liquid crystals in sheet form with an adhesive backing under stock items R20C5W, R25C5W, R29C5W, R30C5W, R35C5W, R40C5W and R35C1W. Other types of liquid crystals which may be useful for the present invention are mentioned in WO 91/09106 (El-Nokaly et al.), U.S. Patent 5,705,093 (Coates et al.) and U.S. Patent 5,690,857 (Osterried et al.). Amounts of the liquid crystal in strips of the present invention may range from about 0.00001 to about 20%, preferably from about 0.01 to about 10%, optimally about 0.5 to about 5% by weight of the strip.

20

Liquid crystal thermochromic substances of the present invention will be capable of operating across many temperature change windows. Some windows may be as small as 0.5° C while others may range as high as 20°C.

25

Flexible water-insoluble substrates are another essential element of the present invention. By "water insoluble" is meant that the substrate does not dissolve in or readily break apart upon immersion in water. A wide variety of materials can be used as the substrate.

30

- 6 -

Nonlimiting examples of suitable substrates include nonwoven substrates, woven substrates, hydroentangled substrates, air entangled substrates and the like. Preferred embodiments  
5 employ nonwoven substrates since they are economical and readily available in a variety of materials. By nonwoven is meant that the layer is comprised of fibers which are not woven into a fabric but rather are formed into a sheet, particularly a tissue. The fibers can either be random  
10 (i.e., randomly aligned) or they can be carded (i.e. combed to be oriented in primarily one direction). Furthermore, the nonwoven substrate can be composed of a combination of layers of random and carded fibers.

15 Nonwoven substrates may be comprised of a variety of materials both natural and synthetic. By natural is meant that the materials are derived from plants, animals, insects or byproducts. By synthetic is meant that the materials are obtained primarily from various man-made materials or from  
20 material that is usually a fibrous web comprising any of the common synthetic or natural textile-length fibers, or mixtures thereof.

Nonlimiting examples of natural materials useful in the  
25 present invention are silk fibers, keratin fibers and cellulosic fibers. Nonlimiting examples of keratin fibers include those selected from the group consisting of wool fibers, camel hair fibers, and the like. Nonlimiting examples of cellulosic fibers include those selected from  
30 the group consisting of wood pulp fibers, cotton fibers,

- 7 -

hemp fibers, jute fibers, flax fibers, and mixtures thereof.  
Wood pulp fibers are preferred.

Nonlimiting examples of synthetic materials useful in the  
5 present invention include those selected from the group  
consisting of acetate fibers, acrylic fibers, cellulose  
ester fibers, modacrylic fibers, polyamide fibers, polyester  
fibers, polyolefin fibers, polyvinyl alcohol fibers, rayon  
fibers and mixtures thereof. Examples of some of these  
10 synthetic materials include acrylics such as Acrilan<sup>®</sup>,  
Creslan<sup>®</sup>, and the acrylonitrile-based fiber, Orlon<sup>®</sup>;  
cellulose ester fibers such as cellulose acetate, Arnel<sup>®</sup>,  
and Acele<sup>®</sup>; polyamides such as Nylons (e.g., Nylon 6, Nylon  
66, Nylon 610 and the like); polyesters such as Fortrel<sup>®</sup>,  
15 Kodel<sup>®</sup>, and the polyethylene terephthalate fibers, Dacron<sup>®</sup>;  
polyolefins such as polypropylene, polyethylene; polyvinyl  
acetate fibers and mixtures thereof.

Nonwoven substrates made from natural materials consist of  
20 webs or sheets most commonly formed on a fine wire screen  
from a liquid suspension of the fibers.

Nonwoven substrates made from synthetic materials useful in  
the present invention can be obtained from a wide variety of  
25 commercial sources. Nonlimiting examples of suitable  
nonwoven layer materials useful herein include HEF 40-047,  
an apertured hydroentangled material containing about 50%  
rayon and 50% polyester, and having a basis weight of about  
43 grams per square yard (gsy), available from Veratec,

- 8 -

Inc., Walpole, MA; HEF 140-102, an apertured hydroentangled material containing about 50% rayon and 50% polyester, and having a basis weight of about 56 gsy, available from Veratec, Inc., Walpole, MA; Novenet<sup>®</sup> 149-191, a thermo-  
5 bonded grid patterned material containing about 69% rayon, about 25% polypropylene, and about 6% cotton, and having a basis weight of about 100 gsy, available from Veratec, Inc., Walpole, MA; HEF Nubtex<sup>®</sup> 149-801, a nubbed, apertured hydroentangled material, containing about 100% polyester,  
10 and having a basis weight of about 70 gsy, available from Veratec, Inc. Walpole, MA; Keybak<sup>®</sup> 951V, a dry formed apertured material, containing about 75% rayon, about 25% acrylic fibers, and having a basis weight of about 43 gsy, available from Chicopee Corporation, New Brunswick, NJ;  
15 Keybak<sup>®</sup> 1368, an apertured material, containing about 75% rayon, about 5% polyester, and having a basis weight of about 39 gsy, available from Chicopee Corporation, New Brunswick, NJ; Duralace<sup>®</sup> 1236, an apertured, hydroentangled material, containing about 100% rayon, and having a basis  
20 weight from about 40 gsy to about 115 gsy, available from Chicopee Corporation, New Brunswick, NJ; Duralace<sup>®</sup> 5904, an apertured, hydroentangled material, containing about 100% polyester, and having a basis weight from about 40 gsy to about 115 gsy, available from Chicopee Corporation, New  
25 Brunswick, NJ; Sontaro<sup>®</sup> 8868, a hydroentangled material, containing about 50% cellulose and about 50% polyester, and having a basis weight of about 60 gsy, available from Dupont Chemical Corp.

- 9 -

Substrates of the present invention need not be formed of fibers. They may be cast as plastic films such as polyethylene, polyester, polyurethane, polyvinyl chloride, polyamide, cellophane, or metallic foils.

5

The water insoluble substrates of the present invention can comprise two or more layers. They may be of similar construction or have different texture and abrasiveness. The differing textures can result from the use of different combinations of materials or from the use of a substrate having a smooth surface and a more abrasive opposite surface. Combinations of hydrophilic and hydrophobic layers may be useful for certain products of this invention. Likewise there may be desired combination of fluid absorptive and non-absorptive layers.

10

15

20

A further important element of cosmetic strips according to the present invention is that of an adhesive composition deposited onto the substrate. The adhesive may be of the pressure sensitive variety or may be a dry-to-the-touch film whose tackiness is generated by adding a small amount of water at the point of use.

25

30

Pressure sensitive adhesives may be formed from a variety of natural and synthetic adhesive polymers. The natural ones may be based on starch or modified starches. Synthetic adhesives include polyvinyl acetate, polyvinyl chloride, polyurethane, polyamide, but most especially acrylic-based polymers. The acrylics may be homo- or co-polymers (the latter indicating at least two different monomer units within the polymer chain). Typical monomers for use in

- 10 -

acrylic-based polymers include acrylic acid, methacrylic acid, ethylacrylate, methylacrylate, butylacrylate and combinations thereof. These may be obtained under the trademark Gelva® series sold by Monsanto and the Duro-Tak® series sold by the National Starch and Chemical Company. Most preferred are acrylic polymers available from Lohmann Therapie Systeme, Germany. Silicone-based polymers may also be employed such as Bio-Psa silicones sold by the Dow Corning Corporation.

10

Release or backing liners are usually placed over the adhesive layer. The release liner should have a surface that is easily stripped off or released prior to use of the strip. Suitable materials for this liner include

15

polyvinylchloride, polyester, polyvinylidene chloride, polystyrene, polyethylene and paper, all of which are preferably but not necessarily coated with a silicone formulation.

20

Another type of adhesive suitable for the present invention is a dry-to-the-touch substance. Products based on this technology are often intended as keratotic plug (e.g. blackhead) removers from facial pores. In a dry state, the adhesive composition is non-tacky to the touch. The

25

adhesive is activated by either directly wetting the composition on the sheet or indirectly by wetting the face in areas to be contacted by the composition. In either instance, the wetting agent interacts with the adhesive composition so it becomes tacky and sufficiently mobile to flow into skin pores. Pure water is the preferred wetting agent. However, other fluid systems or gels could be

30

- 11 -

employed. Suitable fluids would include alcohols such as ethanol, propanol, propylene glycol, polyethylene glycol, polypropylene glycol and especially mixtures of these alcohols with water. Gels would normally consist of fluid  
5 (particularly water) and structuring agents such as Carbomer.

Subsequent to wetting, the adhesive composition is allowed to dry over the area of treatment. During drying keratotic  
10 plugs stickingly adhere to the composition. Advantageously the drying period ranges from 1 minute to 5 hours, preferably from 5 minutes to 1 hour, optimally from 10 to 20 minutes. Thereafter, the dried composition with adhered  
15 plugs is peeled from the skin.

The dry-to-the-touch adhesive composition will include a polymer which may either be anionic, nonionic, cationic, amphoteric or mixtures thereof. Further, there may be utilized combinations of different polymers from within the  
20 same type. Examples of nonionic polymers suitable for film deposition are the copolymers of vinyl acetate and crotonic acid, terpolymers of vinyl acetate, crotonic acid and a vinyl ester of an alpha-branched saturated aliphatic monocarboxylic acid such as vinyl neodecanoate; copolymers  
25 of methyl vinyl ether and maleic anhydride (molar ratio about 1.1) wherein such copolymers are 50% esterified with a saturated alcohol containing from 1 to 4 carbon atoms such as ethanol or butanol; and acrylic copolymers, terpolymers, etc., containing acrylic acid or methacrylic acid, esters of  
30 acrylic or methacrylic acid with one or more saturated alcohols having from 1 to 22 carbon atoms such as methyl

- 12 -

methacrylate, ethyl acrylate, ethyl methacrylate, n-butyl acrylate, t-butyl acrylate, t-butyl methacrylate, n-butyl methacrylate, n-hexyl acrylate, n-octyl acrylate, lauryl methacrylate and behenyl acrylate, glycols having from 1 to 5 6 carbon atoms such as hydroxypropyl methacrylate and hydroxyethyl acrylate, styrene, vinyl caprolactam, vinyl acetate, acrylamide, alkyl acrylamides and methacrylamides having 1 to 8 carbon atoms in the alkyl group such as methacrylamide, t-butyl acrylamide and n-octyl acrylamide, 10 and other compatible unsaturated monomers. One specific example is the emulsion polymerized terpolymer of methacrylic acid, n-butyl acrylate and ethyl acrylate (e.g., in a weight percent ratio of 31:42:27, respectively).

15 Further examples of nonionic film forming adhesive polymers are homopolymers of N-vinylpyrrolidone and copolymers of N-vinylpyrrolidone with compatible nonionic monomers such as vinyl acetate and terpolymers of ethyl acrylate, butyl methacrylate and methyl methacrylate. Nonionic polymers 20 containing N-vinylpyrrolidone in various weight average molecular weights are available commercially from ISP Corporation such as homopolymers of N-vinylpyrrolidone having an average molecular weight of about 630,000 under the trademark PVP K-90 and those having an average molecular 25 weight of about 1,000,000 sold under the trademark of PVP K-120. Particularly preferred is poly(methyl vinyl ether/maleic anhydride) as an unneutralized resin available from ISP Corporation under the trademark Gantrez® S-97 BF and polyvinylformamide available from the National Starch 30 and Chemical Company, Division of ICI.

- 13 -

Anionic film forming adhesive polymers often are derived from the nonionic types which include carboxylic acid functions. Alkaline agents are employed to neutralize the carboxylic acid or anhydride transforming them into anionic salts. Examples of suitable neutralizing agents include

5 2-amino-2-methyl-1,3-propanediol (AMPD);  
2-amino-2-ethyl-1,3-propanediol (AEPD);  
2-amino-2-methyl-1-propanol (AMP); 2-amino-1-butanol (AB);  
monoethanolamine (MEA); diethanolamine (DEA);  
10 triethanolamine (TEA); monoisopropanolamine (MIPA);  
diisopropanol-amine (DIPA); triisopropanolamine (TIPA); and  
dimethyl stearamine (DMS). Most preferred is AMP.

Particularly preferred anionic polymers are the salts of

15 poly(methyl vinyl ether/maleic anhydride) and polystyrene sulfonic acid. The former is obtained by at least partial neutralization of Gantrez® S-97 BF and the latter available from the National Starch & Chemical Company under the trademarks Versa TL-501 and Flexan® 130 having respective

20 molecular weights of about 500,000 and 100,000. Other polymer films which may be employed and are commercially available are listed in the Table below.

TABLE

POLYMER TRADEMARKS (SUPPLIER)	CTFA DESIGNATIONS
Resyn® 28-1310 (NSC)	Vinyl acetate/crotonic acid copolymer
Resyn® 28-2930 (NSC)	Vinyl acetate/crotonic acid/vinyl neodecanoate copolymer
Resyn® 28-2913 (NSC)	Vinyl acetate/crotonic acid/vinyl neodecanoate copolymer
Versatyl® 40 (NSC)	Octylacrylamide/acrylates copolymer
Versatyl® 42 (NSC)	Octylacrylamide/acrylates copolymer
Experimental Resin (NSC)	Vinyl acetate/vinyl neodecanoate/maleic half-ester
Ultrahold-8® (BASF)	Acrylate/acrylamide copolymer
Luviset® CAP (BASF)	Vinyl acetate/crotonic acid/vinyl propionate copolymer
PVP K-30 (ISP)	PVP
PVP/VA E-335 (ISP)	PVP/Vinyl acetate copolymer
PVP/VA E-735 (ISP)	PVP/Vinyl acetate copolymer
Gantrez® ES-225 (ISP)	Ethyl ester of PVM/MA copolymer
Gantrez® ES-425 (ISP)	Butyl ester of PVM/MA copolymer
Gaffix® VC-713 (ISP)	Vinyl caprolactam/PVP/dimethyl aminoethyl methacrylate copolymer

- 5 Cationic adhesive polymers suitable for the present invention may be prepared as homo- or copolymers from monomers including:

- 15 -

Dimethyl aminoethyl acrylate (DMAEA), Dimethylaminoethyl methacrylate (DMAEMA), Dimethylaminopropylacrylamide (DMAPAAm), and Dimethylaminopropyl methacrylamide (DMAPMAAm) which are all (meth)acrylamides or (meth)acrylic acid esters  
5 having a dialkylamino group;

Dimethylaminostyrene (DMAS<sub>t</sub>) and Dimethylaminomethylstyrene (DMAM<sub>St</sub>) and the like which are styrenes having a dialkylamino group;

10

4-Vinyl pyridine and 2-vinyl pyridine which are vinyl pyridines; and

Quaternized products of these with a known quaternizing  
15 agent such as alkyl halide, benzyl halide, alkyl or aryl sulfonic acid, or dialkyl sulfate.

Among suitable amphoteric adhesive polymers are those derived from monomers such as:

20

N-(3-sulfopropyl)-N-acryloyloxyethyl-N,N-dimethylammonium betaine, N-(3-sulfopropyl)-N-methacroylamidepropyl-N,N-dimethylammonium betaine, N-(3-carboxymethyl)-N-methacroylamidepropyl-N,N-dimethylammonium betaine and N-  
25 carboxymethyl-N-methacroyloxyethyl-N,N-dimethylammonium betaine.

When the salt forming group of the cationic and amphoteric polymers is not ionized, it is preferred to ionize it via  
30 neutralization with known acids such as hydrochloric acid and sulfuric acid which are inorganic acids; acetic acid,

- 16 -

propionic acid, lactic acid, succinic acid, glycol acid which are organic acids, or with known bases such as triethylamine, trimethylamine which are tertiary amines; ammonia; or sodium hydroxide.

5

Relative amounts of water-insoluble substrate to adhesive composition may vary in the relative weight range from about 1,000:1 to about 1:1,000, preferably from about 100:1 to about 1:100, optimally from 20:1 to about 1:20, more  
10 optimally from about 5:1 to about 1:5 by weight.

A further element of the present invention is in an agent interactive with water to generate a change of temperature, the agent being admixed with the adhesive polymer. These  
15 agents should be capable of an exothermic or endothermic temperature jump of at least about 2°C, preferably about 5° to about 30°C, more preferably from about 8° to about 20°C, even more preferably from about 10° to about 15° C.

20 Illustrative of the exothermic reaction inducing agents are anhydrous silica, activated alumina, aluminosilicates (e.g. zeolites) and combinations thereof. Particularly preferred are aluminosilicates such as Zeolite A available from PQ Corporation and Cab-O-Sil® fumed silica available from the  
25 Cabot Corporation. Examples of endothermic agents are ammonium chloride and xylitol. Amounts of these substances may range from about 1 to about 80%, preferably from about 10 to about 60%, optimally from about 15 to about 40% by weight of the adhesive composition. When incorporating a  
30 water sensitive temperature change inducing agent,

- 17 -

precaution should be employed to maintain the strip within packaging (e.g. sealed pouch or packet) that avoids exposure to moisture or at least excessive moisture.

- 5 A variety of skin treatment agents may be formulated with the adhesive compositions. These agents may include moisturizers, preservatives, herbal extracts, vitamins, anti-irritant agents, emulsifiers and keratolytic agents.
- 10 Polyhydric alcohols also known as polyols are the most useful moisturizers. Representative polyols include glycerine, diglycerine, propylene glycol, dipropylene glycol, polypropylene glycol, polyethylene glycol and derivatives thereof, sorbitol, hydroxypropyl sorbitol,
- 15 hexylene glycol, 1,2-butylene glycol, 1,2,6-hexanetriol, isoprene glycol, 2-methyl-1,3-propanediol, ethoxylated glycerol, propoxylated glycerol and mixtures thereof. Amounts of the polyol may range from about 0.1 to about 95%, preferably from about 1 to about 50%, more preferably from
- 20 about 1.5 to about 20%, optimally from about 3 to about 10% by weight of the adhesive composition.

- Preservatives can desirably be incorporated into the adhesive compositions to protect against the growth of
- 25 potentially harmful microorganisms. Suitable preservatives include alkyl esters of para-hydroxybenzoic acid, hydantoin derivatives, propionate salts, and a variety of quaternary ammonium compounds. Cosmetic chemists are familiar with appropriate preservatives and routinely choose them to
- 30 satisfy the preservative challenge test and to provide product stability. Particularly preferred preservatives are

- 18 -

phenoxyethanol, methyl paraben, propyl paraben, imidazolidinyl urea, sodium dehydroacetate and benzyl alcohol. Preservatives are preferably employed in amounts ranging from about 0.01% to about 2% by weight of the composition.

Herbal extracts may include Roman Chamomile, Green Tea, Scullcap, Nettle Root, Swertia Japonica, Fennel, Anise, Arnica, Calandula, Coltsfoot, Cornflower, Elder, Gentian, Hawthorn, Lavender, Linden, Myrrh, Oat, Rose, Sweet Clover, Sandalwood, Vetiver, Tulsi, Kamala, Eucalyptus, St. John's Wort and Aloe Vera extracts. Amount of each of the extracts may range from about 0.00001 to about 1%, preferably from about 0.01 to about 0.5%, optimally from about 0.05 to about 0.2% by weight of the adhesive composition.

Vitamins useful in products of the present invention include Vitamin E Acetate, Vitamin C, Vitamin A Palmitate, Panthenol and any of the Vitamin B complexes. Anti-irritant agents may also be present including those of alpha-bisabolol and potassium glycyhrrizinate, each vitamin or anti-irritant agent being present in amounts ranging from about 0.001 to about 0.5%, preferably from about 0.01 to about 0.1% by weight of the adhesive composition.

25

Emulsifiers may also be incorporated into the cosmetic strips of this invention. These emulsifiers may be anionic, nonionic, cationic, amphoteric and combinations thereof. Useful nonionic type emulsifiers include the C<sub>10</sub>-C<sub>20</sub> fatty alcohol or acid hydrophobes condensed with from 2 to 100 moles of ethylene oxide or propylene oxide per mole of

30

- 19 -

hydrophobe; C<sub>2</sub>-C<sub>10</sub> alkyl substituted phenols condensed with  
from 2 to 20 moles of alkylene oxide; mono- and di-fatty  
acid esters of ethylene glycol; fatty acid monoglyceride;  
sorbitan, mono- and di-C<sub>8</sub>-C<sub>20</sub> fatty acids; block copolymers  
5 (ethylene oxide/propylene oxide); and polyoxyethylene  
sorbitan as well as combinations thereof. Alkyl  
polyglycosides and saccharide fatty amides (e.g. methyl  
gluconamides) are also suitable nonionic emulsifiers.  
Amounts of the emulsifiers may range from about 0.1 to about  
10 30%, preferably from about 0.5 to about 10% by weight of the  
adhesive composition.

Keratolytic agents may also be incorporated into the  
adhesive compositions. Typical of these agents are the  
15 alpha and beta hydroxycarboxylic acids. The alpha-  
hydroxycarboxylic acids include glycolic acid, malic acid,  
lactic acid and mixtures thereof as well as their salts such  
as alkali metal and ammonium salts. The most preferred beta  
hydroxycarboxylic acid is salicylic acid. Amounts of these  
20 keratolytic agents may range from about 0.01 to about 15%,  
preferably from about 1 to about 12% by weight of the  
adhesive composition.

Except in the operating and comparative examples, or where  
25 otherwise explicitly indicated, all numbers in this  
description indicating amounts of material ought to be  
understood as modified by the word "about".

The following examples will more fully illustrate the  
30 embodiments of this invention. All parts, percentages and

- 20 -

proportions referred to herein and in the appended claims are by weight unless otherwise illustrated.

#### EXAMPLE 1

5

A dry-to-the-touch strip is prepared for use in removing keratotic plugs from skin pores. The strip employed is a 70:30 rayon/polyester nonwoven fabric available from Dupont. Poly(methyl vinyl ether maleic anhydride), commercially  
10 available as Gantrez S-97® is employed as the adhesive. The resin is dispersed in water along with Microencapsulated Cholesteryl Ester Carbonate (liquid crystal), titanium dioxide, silica and 2-amino-2-methyl-1-propanol (AMP). The mixture is coated by a knife-over-roll onto the nonwoven  
15 substrate. After coating, the strip and adhesive composition are dried at 75°C in a convection oven. The dried sheet is then cut into small strips. The adhesive composition is dry-to-the-touch and has a composition as listed in Table I.

20

TABLE I

COMPONENT	WEIGHT %
Gantrez S-97®	68.1
Anhydrous Silica	20.0
AMP	7.0
Microencapsulated Cholesteryl Ester Carbonate	3.9
Titanium Dioxide	1.0

- 21 -

EXAMPLE 2

Another dry-to-the-touch strip for removing keratotic plugs from skin pores is prepared employing a polyester/cellulose wet layed nonwoven fabric. Poly(vinylformamide) available from the National Starch and Chemical Company is taken as the adhesive in combination with anhydrous silica, titanium dioxide, Vitamin C (ascorbic acid), glycerin, dimethicone copolyol, methoxypropyl glucamide and disodium EDTA. The mixture is coated by a knife-over-roll onto the nonwoven substrate. After coating, the strip and adhesive composition are dried at 75°C in a convection oven. The dried sheet is then cut into small strips. The adhesive composition is dry-to-the-touch and has a composition as listed in Table II.

A sheet of Hallcrest® R20C5W encapsulated thermochromic liquid crystal is attached through a pressure sensitive adhesive backing onto a surface of the fabric opposite the dry layed adhesive.

TABLE II

COMPONENT	WEIGHT %
Polyvinylformamide	52.0
Anhydrous Silica	42.0
Glycerin	1.5
Dimethicone Copolyol	1.3
Vitamin C	1.0
Titanium Dioxide	1.0
Disodium EDTA	0.8
Methoxypropylglucamide	0.4

- 22 -

Just prior to use, the dry adhesive side of the resultant cosmetic strip is wetted. Anhydrous silica present in the composition reacts with the water generating a substantial exotherm. This heat transfers to the liquid crystal layer causing a change in color.

**EXAMPLE 3**

10 This example illustrates a cationic type dry-to-the-touch strip used for the removal of keratotic plugs from skin pores. The substrate employed is a 100% cellulose non-woven fabric. Components of the adhesive as listed under Table III are combined in methanol with stirring and after full  
15 dispersion of the components coated onto the cellulose substrate. Thereafter the system is dried to remove the methanol solvent. The dried sheet is then cut into small strips.

20 A consumer utilizes the strips by wetting face and then applying the strip to the face. Heat is generated by reaction of water with anhydrous zeolite. The temperature increase causes the microencapsulated liquid crystal to change color thereby signaling the time period necessary for  
25 maturation (drying) of strip over the skin pores. Once dried, the strip is peeled. Plugs adhere to the peel being thereby removed from the pores.

- 23 -

TABLE III

COMPONENT	WEIGHT %
Poly-2-acrylamide-2-methylpropane sulphonate	32.0
Zeolite	45.0
Glycerin	15.0
Perfume	5.0
Polyoxyethylene Hydrogenated Castor Oil (60 EO Adduct)	1.0
Microencapsulated Cholesteryl Ester Carbonate (Hallcrest)	2.0

5 EXAMPLE 4

This example illustrates an embodiment utilizing a pressure sensitive adhesive type of dermal patch. A silicone pressure sensitive adhesive is prepared by condensing at

10 about 115°C a siloxane resin copolymer with hydroxyl terminated polydimethyl siloxane in xylene solution in the presence of anhydrous ammonium. The resultant silicone adhesive in an amount of 55% is combined with 35% finely powered silica, 5% ascorbic acid and 5% Microencapsulated  
15 Cholesteryl Ester Carbonate. The combination is deposited onto a polypropylene film to form a dermal patch. A Mylar® backing sheet is placed over the pressure-sensitive adhesive composition for storage and shipping purposes.

20 Just prior to use, the Mylar® backing is removed. Water is applied to an area of the skin onto which the patch is to be placed. Heat generated by the reaction of water with the zeolite causes the liquid crystal to change color.

- 24 -

**EXAMPLE 5**

Another pressure sensitive adhesive strip is prepared for  
5 topical treatment of acne. A breathable polyurethane film  
(Bertek Medfilm 390) serves as a substrate sheet for an  
adhesive composition. An easy release silicon coated  
polystyrene film is then placed over the adhesive layer.  
The final thickness of the dried matrix is between 75 and  
10 150  $\mu$ m. Circular patches are then cut from the resultant  
multi-layered laminate. The formulation of the adhesive  
composition is outlined under Table IV.

**TABLE IV**

15

COMPONENT	WEIGHT %
Polyacrylic Adhesive Resin	66.0
Sodium Aluminosilicate	30.0
Salicylic Acid	0.6
Glycolic Acid	0.4
Sorbitan Monooleate	1.0
Microencapsulated Cholesteryl Ester Carbonate	1.0
Alpha-bisabolol	0.5
Potassium Glycyrrhizinate	0.5

- 25 -

EXAMPLE 6

Microencapsulated pellets of Cholesteryl Ester Carbonate  
5 (5%) are combined with polypropylene pellets and extruded  
through a plastic extruder to form a sheet of thermocolor  
impregnated plastic substrate. Onto this substrate is  
deposited an acrylic based pressure-sensitive adhesive  
formulated with 2% lactic acid. Over the adhesive layer is  
10 applied a release liner covering the tacky adhesive until  
ready for use when the liner is peeled away.

EXAMPLE 7

15 An aqueous dispersion of cholesteryl ester liquid crystal is  
spread onto a cellulose non-woven substrate sheet. The  
sheet is dried in a convection oven to remove water and have  
the liquid crystal thermochromic agent deposited into the  
cellulose substrate. Thereafter pressure-sensitive acrylic-  
20 based resin is applied. Mixed within the acrylic-based  
resin is 40% of powdered, finely dispersed zeolite and 2%  
salicylic acid. A release liner film is applied over the  
adhesive. When ready for use, the liner is removed, and the  
adhesive strip applied to the face. Over several hours,  
25 perspiration penetrates the adhesive layer, interacts with  
the zeolite and thereby emits a heat of reaction. This  
reaction causes the thermochromic agent impregnated into the  
cellulosic strip to turn from blue to colorless. The color  
change indicates to a user the minimum time required for the  
30 strip to release its active amount of anti-acne salicylic  
acid agent.

- 26 -

The foregoing description and examples illustrate selected  
embodiments of the present invention. In light thereof  
variations and modifications will be suggested to one  
5 skilled in the art, all of which are within the spirit and  
purview of this invention.

- 27 -

## CLAIMS:

1. An adhesive cosmetic strip for placement on skin comprising:

- 5           (i) a flexible water-insoluble substrate;  
          (ii)       an adhesive composition deposited onto  
                  the substrate;  
          (iii)       an agent interactive with water present  
                  in a sufficient quantity to induce a change of  
10           temperature in the strip of at least 2°C; and  
          (iv)       a liquid crystal thermochromic substance  
                  incorporated into at least one of the substrate  
                  or adhesive composition, the thermochromic  
                  substance changing color in response to the  
15           change of temperature.

2. The strip according to claim 1 wherein the adhesive is selected from a dry-to-the-touch or a pressure-sensitive tacky adhesive.

20

3. The strip according to claim 1 wherein the thermochromic substance is incorporated into the substrate.

4. The strip according to claim 1 wherein the  
25 thermochromic substance is incorporated into the adhesive composition.

5. The strip according to claim 1 wherein the adhesive composition further comprises a skin treatment agent.

30

- 28 -

6. The strip according to claim 5 wherein the skin treatment agent is selected from the group consisting of herbal extracts, emulsifiers, Vitamins and keratolytic agents.

5

7. The strip according to claim 6 wherein the keratolytic agent is salicylic acid.

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/06796

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 A61K7/48

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 97 32567 A ( KAO CORP (JP)) 12 September 1997 (1997-09-12) whole document, in particular see on page 19, line 3-11 ---	1-7
Y	WO 93 08793 A (BOOTS CO PLC) 13 May 1993 (1993-05-13) page 2, line 20 - line 28 ---	1-7
Y	GB 1 597 388 A (CELLOGRAF SIMP SPA) 9 September 1981 (1981-09-09) claims ---	1-7
A	FR 2 205 303 A (PIERRET COLETTE) 31 May 1974 (1974-05-31) page 3, line 17 - line 22 claims --- -/-	1



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

### \* Special categories of cited documents:

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*&\* document member of the same patent family

Date of the actual completion of the international search

12 December 2000

Date of mailing of the international search report

20/12/2000

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel (+31-70) 340-2040, Tx. 31 651 epo nl.  
Fax (+31-70) 340-3016

Authorized officer

Pelli Wablat, B

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/06796

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 642 250 A (SPECTOR DONALD) 10 February 1987 (1987-02-10) claims	1
	---	
A	WO 97 48387 A (LAVIPHARM SA (GR)) 24 December 1997 (1997-12-24) claims	1-7
	-----	

# INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern. Application No

PCT/EP 00/06796

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 9732567	A	12-09-1997	JP 2935343 B	16-08-1999
			JP 11012127 A	19-01-1999
			AU 702300 B	18-02-1999
			AU 2231297 A	22-09-1997
			BR 9707907 A	27-07-1999
			CA 2248326 A	12-09-1997
			CN 1216917 A	19-05-1999
			EP 0904049 A	31-03-1999
			US 6042844 A	28-03-2000
WO 9308793	A	13-05-1993	AU 2888292 A	07-06-1993
GB 1597388	A	09-09-1981	BE 862987 A	16-05-1978
			CH 620295 A	14-11-1980
			DE 2801434 A	27-07-1978
			FR 2377885 A	18-08-1978
			US 4220680 A	02-09-1980
FR 2205303	A	31-05-1974	BE 806938 A	01-03-1974
			CA 974884 A	23-09-1975
			CH 572743 A	27-02-1976
			DE 2355074 A	06-06-1974
US 4642250	A	10-02-1987	NONE	
WO 9748387	A	24-12-1997	GR 1002807 B	13-11-1997
			US 5976565 A	02-11-1999
			AU 3340997 A	07-01-1998
			BR 9709920 A	11-01-2000
			CA 2258242 A	24-12-1997
			CN 1226160 A	18-08-1999
			EP 0917461 A	26-05-1999
			FR 2750050 A	26-12-1997
			JP 2000513347 T	10-10-2000
			NO 985938 A	18-02-1999
			PL 330848 A	07-06-1999